

Mercury Levels in Surface Waters of the Carson River/Lahontan Reservoir System, Nevada: Influence of Historic Mining Activities

J. C. Bonzongo^{1*}, K. J. Heim², J. J. Warwick², and W. B. Lyons³

1. Center for Environmental Sciences and Engineering / MS.199 - University of Nevada - Reno, NV 89557- 0014

2. Hydrology and Hydrogeology Program / MS.175 - 1000 Valley Rd, University of Nevada - Reno, NV 89512-0180

3. Department of Geology, University of Alabama, Tuscaloosa, Al 35487-0338

*To whom correspondence should be addressed

The current address of JC. Bonzongo is Department of Geology, University of Alabama, Tuscaloosa, Al 35487-0338.

ABSTRACT

Total mercury (Hg_T), methylmercury (MeHg), and other operationally defined Hg species were determined on water samples collected from a fluvial-lacustrine system impacted by historic mine wastes. Simultaneously, a comprehensive study was undertaken to determine the influence of some major physio-chemical parameters on the fate of Hg within the system. Total Hg levels showed an increase from common background concentrations of 4 ng/L upstream of mining activity, to peak values of 1500-2100 ng/L downstream of Hg contaminated mine tailings piles. Methylmercury concentrations varied from 0.1 to 7 ng/L in oxygenated surface waters. In both cases, peak values were associated to the highest concentrations of total suspended solids (TSS). Particulate total Hg (Hg_P) was typically >50% of Hg_T increasing downstream. In contrast, the dissolved fraction of MeHg (MeHg_D) constituted always a significant portion of total methylmercury (MeHg_T). The $[\text{MeHg}_T]/[\text{Hg}_T]$ ratio decreased downstream suggesting both a high percentage of inorganic Hg input from point sources, and likely low specific rates of MeHg production within the aquatic system. The latter being due to the combined effects on microbial population of both high levels of Hg concentrations found in water and sediments, and other factors related to the aqueous geochemistry of the system. Concentrations of Hg_T in the water column were controlled by the input of contaminated particles from the watershed during the spring snow melt. In Lahontan Reservoir, significant losses of Hg from the water column were observed, especially at Lahontan dam. Two major pathways of Hg removal from the water column were identified: (1) losses of Hg bound to particles by sedimentation, and (2) removal by volatilization of dissolved gaseous Hg (Hg^0) to the atmosphere.

INTRODUCTION

Mercury (Hg) is the most toxic of the common heavy metals which have been identified as pollutants, even at very low concentrations (Largewerff, 1972; WHO, 1990). With no known essential biological function, Hg is bioaccumulated and bioconcentrated. The above aspects of Hg have generated public concerns on its disposal and fate in the environment. During the past decade much of the attention regarding Hg studies has shifted from aquatic systems with direct point sources of Hg, to those systems with poorly defined sources (Wiener and Stokes, 1990). The latter are represented by remotely located low-pH and poorly buffered systems and newly created impoundments (Hakanson, 1980; Gill and Bruland, 1990; Grieb et al., 1990; Verdon et al., 1991). These ecosystems exhibit elevated Hg levels in fish tissues, frequently exceeding public health guidelines (0.5 - 1 µg/g wet weight). In these remote aquatic systems without any known direct discharge point source of pollutants, Hg is thought to be supplied by both direct deposition from the atmosphere and input via runoff from the catchment (Swain et al., 1992; Winfrey and Rudd, 1990; Lee and Hultberg, 1990).

Mercury cycling is of continuing interest in areas receiving a direct influx of Hg from both recent and/or historic anthropogenic activities, the latter of which is the case for the Carson River Drainage Basin (CRDB) in Nevada. Here, many large hard rock mining claims were developed in the latter part of the 19th century. The extraction of precious metals was accomplished by an inefficient and crude amalgamation process, in which liquid Hg was used to free gold and silver from crushed ores. This process introduced about 7,500 tons of Hg to the environment and a significant portion of these losses entered directly or indirectly the Carson River (Smith, 1943). This mining activity resulted in a high Hg contamination level in multiple media of the CRDB (Cooper et al., 1985; Gustin et al., 1994). Consequently, Hg concentrations in the Carson River/Lahontan Reservoir system are much higher than anomalies found in many aquatic systems which have been investigated elsewhere for Hg biogeochemistry in the latter decades. In addition, the Carson River and its tributaries are characterized by: (1) alkaline pH values (7.3-8.9), (2) an evapoconcentration of conservative chemicals resulting from the aridity of the climate, (3) their

location on a natural mercuriferous belt, and (4) their particular hydrogeochemistry (Cooper et al. 1983; Cooper et al., 1985; Johannesson et al., 1992; Doyle et al., 1994; Gustin et al., 1994).

Despite numerous studies on Hg biogeochemistry, limnological factors responsible for its cycling and the occurrence of its different species in freshwater systems are still not well understood (Gill and Bruland, 1990). In addition, these factors may vary widely from one system to another. Among factors known to influencing Hg bioaccumulation in aquatic organism tissues are the concentrations of Hg_T and Hg speciation in water (Meili et al. 1991). Therefore, for a given aquatic system, the investigation of the geochemical behavior and the cycle of predominant species of Hg, as well as the identification of key limnological parameters, may help to better predict both environmental and human health effects.

The CRDB has been contaminated for over a century. However, only few studies have been conducted to determine the extent of Hg contamination and transformation in the aqueous phase. Here, we report results of Hg concentrations and speciation in surface waters from the Carson River/Lahontan Reservoir system. Environmental factors influencing Hg speciation and consequently its bioaccumulation are examined.

STUDY AREA

The area under investigation is a section of the Carson River in western Nevada. This section extends from the upper Carson River near the Nevada State Penitentiary at Carson City, downstream to Lahontan Dam (Fig. 1). The Carson River flows northeast from the Sierra Nevada through Lahontan Reservoir, providing irrigation water to areas upstream of the reservoir. Flows in the Carson River range from little to none during late summer, to well over 15,000 cfs ($424.8 \text{ m}^3/\text{sec.}$) during winter and spring flooding events. The river terminates in the Carson Playa. Lahontan Dam was built in 1915 as a result of the Federal Reclamation Act of 1902 which authorized the construction of the Newlands Irrigation Project. The construction of the Truckee Canal which supplies the reservoir with about 40% of its water annually was also a direct result of the same project. Sources of water for Lahontan Reservoir are the combined drainages of the

Carson and Truckee Rivers, about 3,120 square miles (8085 km²) (Katzner, 1971). The water from the reservoir is utilized during the growing season to irrigate agricultural lands in Lahontan Valley. The reservoir is also managed as a warmwater fishery which is supported primarily by white bass (*Morone chrysops*), white crappie (*Pomoxis annularis*), yellow perch (*Perca flavescens*), white catfish (*Ictalurus catus*), and channel catfish (*Ictalurus punctata*) (Cooper et al. 1983). Prior to the construction of the dam, the water from the Carson River emptied into the Carson Lake which is the natural terminus of the river.

MATERIALS AND METHODS

Containers and cleaning processes. All samples were collected in prewashed Teflon[®] bottles. New bottles were rinsed in NANOpure[®] water and immersed in a warm micro-soap[®] bath for 3 days then rinsed thoroughly with NANOpure[®] water, immersed in 50% HNO₃ at 90 °C for 5 days. After the acid bath, they were rinsed with pure water and filled with a small amount of Optima[®]-HCl 6N (obtained from Fisher Co) for cleaning by reflux for 2 days. This step was repeated. Finally, bottles were rinsed 3 times with NANOpure[®] water before being filled with pure water containing 1% Optima[®]-HCl and double-bagged. For the cleaning of used bottles, only acid leaching steps were used.

Sample collection, storage, and preservation. Samples were collected using the "ultra-clean free-metal sampling" protocol (Gill and Fitzgerald, 1985). Surface water samples were collected during 2 different flow regimes (546 and 63 cfs) at eight stations (Figure 1). These stations were chosen based on their location relative to known Hg contaminated sites and Hg distribution within the river/reservoir system (Cooper et al. 1985). Samples were also collected from the Truckee Canal to determine the influx of Hg from the Truckee River. Clean polyethylene gloves were worn during the collection of samples. Once the acidified water was emptied, bottles were rinsed 3 times with the river water before being filled. Samples for the determination of elemental Hg and the acid-labile Hg were collected in 1 liter Teflon[®] bottles. Samples for Hg_T were collected using 250 ml Teflon[®] bottles. Samples for MeHg determination and splits for Hg_T

were collected in Teflon[®] bottles obtained from Brooks Rand Ltd. or Frontier Geosciences (both in Seattle, WA). After sample collection, and during their transportation to the laboratory, samples were stored in coolers containing ice packs. Samples for the determination of elemental mercury were purged within 5 hours of collection. Filtration (0.45 μ m acid precleaned polycarbonate filters) and acidification (1% v/v with Optima[®]-HCl) were conducted in the laboratory under a controlled atmosphere in an HEPA[®] hood.

Analysis. A nondispersive atomic fluorescence spectrometer (CVAFS) was used for the measurement of Hg in the gas phase. Different forms of Hg were operationally named according to the steps involved in the treatment of the sample before the gas-phase stripping (with high purity He or N₂), and Hg trapping onto a gold-sand trap. Mercury was thermally desorbed from the gold-sand trap and quantified by a CVAFS (Bloom and Crecelius, 1983; Gill and Bruland, 1990).

-*Elemental mercury (Hg^0)* was determined by purging non-acidified fresh water with Hg-free helium. Briefly, ultra-pure helium (grade 5) was passed through a stack containing charcoal and 3 blanked gold-sand traps before reaching the bubbler. The bubbler was a 2 liter Teflon[®] bottle, connected to a line containing; first 1 tenax column for the removal of organic mercury compounds, and second, a blanked gold-sand trap for trapping Hg^0 contained in the sample. One liter of sample was used and purged for 30 minutes under a controlled atmosphere (HEPA hood) in the laboratory.

-*Acid-labile or reactive Hg (Hg_R)* includes Hg species reduced by $SnCl_2$ to Hg^0 , and corresponds to inorganic and organically complexed Hg species that are displaced by protons during acidification of the sample to pH ~1 (Bloom and Crecelius, 1983; Gill and Bruland, 1990). For this determination, 500 ml of nonfiltered acidified sample (1%) is placed in a bubbler and 0.5 ml of $SnCl_2$ (30%) is added. The sample is then purged for 30 minutes on a gold-sand trap separated to the bubbler by a quartz tube containing soda-lime (Bloom and Crecelius, 1983).

-*Methyl mercury.* Total methylmercury ($MeHg_T$) and its dissolved fraction ($MeHg_D$) were analyzed by Frontier Geosciences (Seattle, WA) using distillation and ethylation processes before analysis by CVAFS (Bloom, 1989; Horvat et al., 1993). Only one sample was collected per station

during the spring runoff. In June, three samples were collected at each location for the determination of sample variance.

-*Total mercury (Hg_T) and its dissolved fraction (Hg_{TD})* were determined on field triplicates with $SnCl_2$ reduction technique (Bloom and Crecelius, 1983). Prior to analysis, the aqueous sample (10 to 100 ml) containing 1% Optima[®]-HCl was oxidized in a 125 ml Teflon[®] bottle by adding 0.5 ml of $BrCl$ per 100 ml of sample. Samples containing high levels of Hg were run on small volumes on purged NANOpure water.

-*Hg detection limit and precision:* The detection limit (defined as 3 times the standard deviation of the average purge blank, 0.028 ± 0.01 , $n=6$) for the analysis of an aqueous sample was 0.03 ng or 0.1 ng/L for a 200 ml water sample. The precision on a 1 ng standard solution prepared using the atomic absorption Hg standard was 1% (1.067 ± 0.015 ; average \pm one standard deviation, $n=10$). The coefficient of variation based on 8 analyses of one sample collected at the Carson River Delta was 5% (922 ± 50 ; average \pm one standard deviation). The percentage of recoveries on spiked samples ranged from 98 to 106%. In addition to procedural and field blanks consisting of acidified NANOpure[®] water, the quality assurance and quality control purposes were met by sending duplicate water samples to Brooks Rand Ltd. or Frontier Geosciences (Seattle, WA). For samples collected from uncontaminated sites (locations 1 and 2, Figure 1.), variations in Hg concentrations with our laboratory at the University of Nevada, Reno ranged from 1 to 5% for both Hg_T and Hg_{TD} . However, filtered samples collected in May at Fort Churchill and Fisherman's Point showed a broad range of values.

Other parameters. During the collection of water samples for Hg analysis, some parameters were measured in situ (Dissolved oxygen, pH, and temperature). Samples were also collected in polyethylene bottles for the determination of chemical variables influencing Hg cycling. Methods for chemical analyses of water and waste water (EPA), were used for the determination of: chloride (Cl^-), sulfate (SO_4^{2-}), dissolved organic carbon (DOC), and hydrogen sulfide (H_2S). These variables were determined at the Water Resource Center of the Desert Research Institute (Reno, NV). Total suspended solids (TSS) were determined on a 1 liter sample

volume, filtered through a 0.45 μm pre-weighed filter, and dried 24 hours at 60 $^{\circ}\text{C}$.

Data analysis. Statistical analyses were conducted with SAS[®] (SAS Institute, 1985). Statistical methods, such as linear and PROC STEPWISE multiple regression analyses were used to examine the influence of key limnological parameters on the fate of Hg within the Carson River/Lahontan Reservoir system. The parameters tested were chemical variables describing water quality: dissolved oxygen, pH, temperature, dissolved organic carbon, sulfate, and chloride. Three dependent variables, total-Hg, methylmercury, and total acid-labile Hg, were subjected to correlation and multiple regression analyses.

RESULTS and DISCUSSION

Mercury concentrations and speciation. The distribution of Hg_T in water in the Carson River/Lahontan Reservoir system shows the existence of three distinct regions. The first region is located upstream of the historic mill sites where Hg levels are relatively low, representing the local background levels (stations 1 and 2, Figure 1). The second occurs, from Dayton to the Carson River delta (stations 3 to 6), where Hg contamination occurs from mine tailings spatially distributed in the floodplain, channel banks, and bed sediments. The third region is the artificial terminal sink (reservoir) where an important sedimentation process occurs, allowing the reduction of turbidity and Hg concentrations observed in water downstream of Lahontan dam (Cooper et al. 1985).

The range of mean values of Hg_T concentrations determined in the two surveys was broad (4.28 - 2107 ng/L) and showed the impact of mine tailings spatially distributed along the river (Table 1). During the spring runoff in May, the peak value of Hg_T was observed at Fort Churchill. One month later, when the water discharge was about 15% of what it was in May, the peak was shifted downstream to Fisherman's Point in the reservoir. In both cases, Hg_T peak values matched the highest concentrations of TSS (Tables 1 and 2). This suggests that Hg concentrations in water were primarily controlled by inputs of Hg-contaminated particles eroded from the watershed and

the river banks during the peak flow period. This observation is in accord with the positive relationship ($r=0.90$, $p<0.01$) between Hg_T concentrations and water discharges, observed previously in the Carson River (Cooper et al., 1985). The input of Hg-contaminated particles can also explain the relatively high concentrations (12.88 and 28.15 ng/L) determined on samples collected upstream of Dayton in May. In general, concentrations of Hg_T steadily increased from Carson City downstream to the Carson River Delta in May, and to Fisherman's point in June, showing a similar range and trend to those observed in previous investigations (Cooper et al., 1985; Wayne et al. 1994).

In the reservoir, the highest concentrations of Hg_T and TSS were observed during the lowest flow regime when inputs from the watershed were negligible. This increase in Hg_T concentrations in the river from Carson City Gage, downstream to Fisherman's Point in the reservoir, is probably related to the size fractionation and the nature of TSS. While moving downstream, the finest grained particles which have high adsorption capacity for trace metals (McCutcheon et al., 1992), remain longer in suspension than the coarse fraction. From Fisherman's Point to Lahontan dam, a significant decrease was observed in Hg concentrations. Total mercury concentrations were 10 and 22 times higher at Fisherman's point than at Lahontan dam in June and May respectively (Table 1). Approximately, 90% to 95% of Hg_T found at Fisherman's Point was removed from the water column before reaching the dam site while the concentrations of chloride, the conservative tracer, were increasing. This observation points out the importance of Lahontan Reservoir as a Hg sink. Because of the long hydraulic detention time of Lahontan Reservoir which has been estimated to be about 9 months, and the production of biogenic materials which takes place there at the end of winter (Cooper et al., 1983), the main removal mechanism of Hg is likely linked to its adsorption on suspended particle matters followed by subsequent flocculation and sedimentation. In contrast, because of the input of cleaner water from the Truckee Canal (Figure 1) with an average Hg_T concentration of 4 ng/L, no significant differences were observed in TSS concentrations between stations 7 and 8 in the reservoir (Table 2).

The range of mean total dissolved mercury (Hg_{TD}) concentrations varied from 2.26 to 56.46

ng/L, and showed a similar trend to that of Hg_T in June. However, in May, mean Hg_{TD} values were surprisingly high at stations 1, 5, and 7. At stations 1 and 2, these Hg_{TD} values were associated with high TSS concentrations; 50 and 73 mg/L respectively, but only 45% of Hg_T was bound to the solid phase at the Hg background level station (figure 3). That was due to the composition of TSS at this particular location, where sand materials which do not have high affinity for Hg (Akagi et al. 1979), represented the highest fraction.

Mean concentrations of MeHg ranged from 0.3 to 7.2 ng/L, and from 0.1 to 1.6 ng/L for total and the dissolved fraction respectively. The absolute values of MeHg_T were higher than that of many previously documented MeHg concentrations in oxic freshwaters with or without sources of Hg. However, the $[\text{MeHg}_T]/[\text{Hg}_T]$ ratio decreased consistently downstream, reaching values less than 1% of Hg_T in stations located downstream of the contaminated mine tailings. The highest percentages of MeHg_T (6.4 - 12%) were found upstream of the contaminated section. These observations suggest both a small proportion of MeHg_T versus Hg_T from Hg-discharge points and probably a low net product of methylation/demethylation within the system. The reason for the latter observation may be twofold. (1): Since the studied aquatic system has been heavily contaminated by Hg for over a century, microbial communities have had long time to respond to Hg stress. Accordingly, established detoxification mechanisms which involve specific enzymes, can increase the demethylation process, affecting the accumulation of MeHg (Barkay, 1987; Baldi et al., 1989), and (2): Sulfate reducing bacteria (SRB) have been identified as principal methylators of Hg in both estuarine and freshwater sediments (Compeau and Bartha, 1985; Gilmour and Henry, 1992). Their activity with regards to MeHg production can be affected by both low (<200 μmoles) and high (>500 μmoles) levels of sulfate concentrations, as well as by molybdate, a known inhibitor of sulfate metabolism by SRB (Gilmour and Henry, 1991). The occurrence of these two compounds in relatively high concentrations in the Carson River/Lahontan Reservoir system, may impact MeHg production by SRB. Concentrations of molybdenum have an inverse trend relative to that of $[\text{MeHg}_T]/[\text{Hg}_T]$ ratio in the Carson River (W.B. Lyons, pers. communication). In addition, sulfide, the end product of sulfate reduction was always under the

analytical detection limit (Table 2), even in bottom anoxic waters of the reservoir, collected in late summer (Bonzongo et al. 1994).

The particulate fraction (Hg_p) always constituted a significant portion of Hg_T concentrations, reaching 94 to 99% from Fort Churchill downstream Lahontan dam. In contrast, the dissolved fraction of MeHg ($MeHg_D$), even decreasing downstream was always significant (Figure 2). Coefficients of distribution (K_d) of Hg_T and $MeHg_T$ between the solid and dissolved phases were determined using the following equation: $\log K_d = \log(C_s/C_w)$, where C_s and C_w are concentrations of Hg_T or $MeHg_T$ in particulate matters (ng Hg/kg) and in water (ng Hg/L) respectively. The obtained K_d varied from 4.2 to 6.7 for Hg_T , and 4.0 to 5.5 for $MeHg_T$. The partitioning of both Hg_T and $MeHg_T$ was weakly related to pH and concentrations of chloride. Coefficients of distribution of MeHg between the dissolved phase and seston in the studied system were higher than K_d values reported for sand, silt and woodchips, woodchips, and DOC (<4.0) (Akagi et al., 1979; Miskimmin, 1991). However, they were quite similar to K_d values reported for seston in remote seepage lakes (Bloom et al., 1991). For Hg_T K_d values were higher than those given for lake sediments (Hesslein et al., 1987). Since the higher the value of K_d , the lower the dissolved Hg fraction, these values indicate that total suspended solids play an important role in the fate of Hg in the Carson River/Lahontan Reservoir system. Brought in the system mainly via runoff during the snow melt period, they are vectors of Hg in the water. They control the transport of Hg downstream from discharge points and its removal by sedimentation. Therefore, TSS influences the accumulation of Hg in aquatic biota by reducing its availability for conversion processes and uptake.

The acid-labile Hg, which represents the Hg substrate available for methylation, Hg^0 formation, and other conversion processes (Mason and Fitzgerald, 1990) constituted always a small fraction of Hg_T . The peak value of Hg_R (12.88 ng Hg/L) was recorded in the reservoir at Fisherman's Point in June. This value was far above the range of Hg_R concentrations determined during the two surveys (0.4-4.0 ng Hg/L). This fraction had a positive relationship with Hg_T ($r=0.65$, $p=0.02$), and a similar trend with that of TSS concentrations. The "nonreactive" fraction of Hg

defined here as the difference between Hg_T and Hg_R represented 90 to 99.9%. This indicates that only a very small fraction of Hg found in the water is available for conversion processes (Mason and Fitzgerald, 1990).

Elemental Hg was determined on samples collected in June. Its concentrations were around 0.1 ng/L upstream of Fort Churchill and averaged 1 ng/L downstream. The determination method used here discriminates between dissolved gaseous organic Hg and dissolved Hg^0 . From previous studies, it is assumed that all the dissolved gaseous Hg in surface water and all the inorganic gaseous Hg in the overlying air is Hg^0 (Kim and Fitzgerald, 1986; Fitzgerald et al., 1991; Vandal et al., 1991). A recent investigation in the CRDB has shown that the range of concentrations of Hg in the atmosphere was broad and varied from common background concentrations (1 to 4 ng/m³) to values as high as 240 ng/m³ over contaminated sites (Gustin et al., 1994). In the Lahontan Reservoir area, where substantial concentrations of Hg^0 were detected in water, the atmospheric concentration averaged 2.3 ng/m³ (Gustin et al., 1994). This value was used to estimate the degree of saturation for Hg^0 in the reservoir water as described by Vandal et al. (1991), using the following equation: $S = [(C_w * H) / C_a] \times 100$, where C_w and C_a are the concentration of Hg^0 in water and air respectively, H, the Henry's law constant for Hg^0 , and S, the saturation percentage. As previously observed in the equatorial Pacific Ocean (Kim and Fitzgerald, 1986), in temperate lakes in Wisconsin (Vandal et al. 1991), and in Pettaquamscutt estuary (Mason et al., 1993), surface water in Lahontan Reservoir was supersaturated with Hg^0 relative to the atmosphere concentration ($S > 100$). This indicates the existence of losses of Hg from the water column to the atmosphere by volatilization. Because of the use of metallic Hg in the amalgamation process, Hg^0 in the river section is introduced in the water from contaminated mine tailings which are distributed between Dayton and the mouth of the reservoir (Figure 1). This observation was confirmed by the results of Hg speciation in sediment, which showed high levels of Hg^0 near mine tailings while in the reservoir its concentrations were below the analytical detection limit (Miller et al., 1994). Since the solubility of Hg^0 in water is not high enough (Lindqvist et al., 1984), the increase of its concentrations in the reservoir after the removal of more

than 90% of the Hg-load suggests an *in situ* production, especially at the dam site. If so, that production is likely the result of biological activity involving: (1) bacteria able to produce specific enzymes for both Hg-demethylation and reduction of Hg^{2+} ions to Hg^0 (Barkay, 1987; Baldi et al., 1989), and (2) phytoplankton (Vandal et al., 1991; Mason et al. 1993). Lahontan Reservoir being eutrophic (Cooper et al., 1983), the effect of phytoplankton can be related to the generation of humic substances which can act abiotically, reducing Hg^{2+} ions to Hg^0 (Jackson, 1988; Allard and Arsenie, 1991).

Overall, two major pathways are involved in the self-cleaning capacity of the water in Lahontan Reservoir: (1) removal of Hg scavenged on particles by sedimentation, and (2) losses by volatilization of dissolved gaseous Hg to the atmosphere.

Comparison with other recent reports on Hg in freshwaters. The coexistence of polluted and nonpolluted areas in the studied system gives an opportunity for comparison with data obtained elsewhere, from both Hg-impacted and noncontaminated aquatic systems. A comparison of Hg data obtained in the present study and that found in recent literature, is given in table 3. Davis Creek and Clear Lake, located in Central California, were selected for comparison because they have some similarities with the Carson River/Lahontan Reservoir system, including: high pH values (7.5-9), a similar range of dissolved organic carbon (3-6 mg/L), and Hg pollution related to mining activities (Cooper et al. 1983; Cooper et al., 1985; Gill and Bruland, 1990; Gustin et al., 1994). In contrast, Clay Lake (Ontario, Canada) and Onondaga Lake (NY) were mainly polluted by chlor-alkali plant discharges, the latter, with epilimnion pH ranging from 7.5 to 8.3 has been described recently as one of the most polluted lakes in the United States (Bloom and Effler, 1990). Also listed are Lake San Antonio (CA) and Pyramid Lake (NV) which lack direct sources of Hg contamination.

Despite the fact that ranges of data for Onondaga lake include values determined on anoxic bottom water samples, both Hg_T and Hg_{TD} concentrations in the listed aquatic systems are relatively close to, or included in, the range of Hg concentrations found in the background area of

the the Carson River/Lahontan Reservoir system. As shown in table 3, Hg_T and Hg_{TD} concentrations in the contaminated part of the Carson River are 5 to 100 times higher than those given in the selected references, and likely among the highest in aquatic systems with known histories of Hg contamination.

In contrast to large differences in Hg_T concentrations, patterns of Hg speciation are more comparable. The highest concentration of MeHg_T in oxic unfiltered surface water of the Carson River was approximately one order of magnitude higher than those observed in Clay and Onondaga lakes. This peak value was similar to that obtained in anoxic bottom water of Onondaga Lake. The dissolved fraction of MeHg in Onondaga Lake was in the range of concentrations found in regions 1 and 3 of the Carson River/Lahontan Reservoir system. However, concentrations of MeHg_D in region 2 were much lower than those observed in anoxic water in Onondaga Lake. Overall, in non and less contaminated systems in Central California, MeHg_D represented a significantly higher fraction of Hg_{TD} compared to those observed in the Carson River. This observation might be related to the difference in the speciation procedure.

The acid-labile fraction in the Carson River varied from 1% to 11% of Hg_T . However, it concentrations represented 4.5% to 57% of Hg_T in surface water of the Onondaga Lake and up to 50% of Hg_T in nonpolluted systems. These differences are likely due to the nature and abundance of materials (sand, silt, clay, humic substances, bacteria, and plankton) which constitute the TS in these aquatic systems.

Mercury and environmental factors. The distribution and the occurrence of Hg species in aquatic environments are probably regulated by some key limnological and chemical parameters even though little is known about the relation between these factors and Hg speciation (Gill and Bruland, 1990; Bloom and Effler, 1990). Using the combined data obtained from the two sampling events, we examine the influence of physiochemical parameters on concentrations and speciation of Hg. Because of its dissimilarity, regression analyses were run with and without data from Lahontan dam. In the latter case, positive and significant relationships were obtained between

Hg and the distance downstream from Carson City, and TSS (table 4). When data from Lahontan dam were included, significant relationships were restricted to correlations between Hg_T , Hg_{Me} and TSS. In contrast, the percentage of MeHg (not shown) for unfiltered samples showed negative but significant relationship with the distance downstream from Carson City ($r=-0.7$, $p=0.01$). This relation suggests both the importance of the inorganic fraction of Hg from point sources and likely a low specific rate of net MeHg production in the aquatic system. For filtered samples, a similar relation was found with TSS ($r=-0.84$, $p=0.001$), showing the high affinity of Hg to particles. As mentioned above, this affinity is responsible for both longitudinal and vertical transport of Hg bound to particles.

If Hg concentrations were increasing downstream and showing a positive relation with TSS, then other parameters were not significantly correlated with Hg concentrations. However, in natural environments, Hg cycling is more likely under the combined influence of many parameters. These relationships can be described by multilinear equations which can be used to predict levels of Hg in surface water. To find regression equations containing two to three independent variables accounting for >80% of the variation of the dependent variables, concentrations of Hg_T and that of different Hg species were used as dependent variables in stepwise regression analyses, data from Lahontan dam (station 8) being excluded. For each dependent variable, eight independent variables or more were tested.

For Hg_T , the best model from the stepwise regression procedure had the distance downstream and TSS as independent variables, accounting for 86% of the variation in Hg_T concentrations (adjusted $R^2 = 0.86$; $p = 0.0003$), yielding the equation:

$$Hg_T(\text{ng/L}) = -412 + 13.22[\text{Distance downstream (Km)}] + 14.31[\text{TSS (mg/L)}] \quad (1)$$

Methylmercury varied with Hg_T in a positive and significant relationship ($R^2=0.86$; $p = 0.0003$) with the following equation:

$$\text{MeHg}_T (\text{ng Hg/L}) = 0.55 + 0.0026 [\text{Hg}_T (\text{ng/L})] \quad (2)$$

For MeHg_D and Hg_R , the best n -variable models included more than four independent variables. No significant relationship was obtained with the dissolved fraction of Hg_T .

From these analyses, only Hg_T and MeHg_T may be fairly predictable using equations (1) and (2), if the sampling point as well as TSS concentrations are known.

To investigate issues related to MeHg production and consequently the accumulation of Hg in biota, Hg partitioning as well as the relationship between its dissolved fractions and DOC are of importance. Concentrations of DOC in the Carson River/Lahontan Reservoir system range from 3 to 6.4 mg/L (Table 2). However, these concentrations were poorly and negatively correlated to dissolved fractions of Hg (Table 4), indicating a poor complexation of Hg_{TD} to dissolved organic ligands. Accordingly, it can be assumed that a significant fraction of Hg_{TD} in the studied system is available for conversion processes and direct uptake via diffusion through biotic boundaries. The simplest way for the determination of the fraction which remains in solution when concentrations of TSS are known is given by following relationship (McCutcheon et al 1992): $F_s = 1 / [1 + (K_p * \text{TSS})]$, where F_s is the ratio of dissolved to total concentrations, K_p , the distribution coefficient (L/Kg), and TSS, total suspended solids (Kg/L).

When data from stations 1 (outside of the contaminated area) and 8 (affected by the Truckee Canal) were excluded because of their dissimilarities, the K_p obtained from the above equation which yielded the best fit regression curves were 1.2207×10^6 and 7.6×10^4 L/kg for Hg_T ($r^2=0.92$) and MeHg_T ($r^2=0.56$) respectively. Accordingly, these K_p can be used to predict distribution of Hg between the dissolved and sorbed phases in the studied system.

CONCLUSION

The Carson River/Lahontan Reservoir system is one of the most heavily Hg -contaminated water bodies in the United States. The intensive annual erosion of contaminated particles which occurs in conjunction with snowmelt and associated runoff, constitutes the main factor leading to the Hg

load in the system. Within the fluvial-lacustrine system, the fate of Hg remains in large part linked to that of suspended and settling particles. Trends of Hg concentrations recorded during the two sampling events showed that MeHg was significantly brought in the system from external discharge point sources. This observation was confirmed by the good correlation ($r=0.99$) between concentrations of MeHg in the water and in river bank materials (Bonzongo et al., in preparation). Rates of the *in situ* production of MeHg during the sampling periods were probably low, especially in the river section, or affected by the demethylation process, due to the activity of Hg-resistant microbial communities.

The reservoir behaves as a terminal sink. In this part of the system, an important removal of Hg is associated to the sedimentation process taking place. The supersaturation of surface waters with Hg^0 observed in the reservoir indicates that the atmosphere is also a sink of Hg lost by volatilization.

In late summer, when the water input from the Carson River is negligible, the reservoir becomes thermally stratified with anoxic hypolimnion in its deepest location. In that period, an algal bloom is observed in conjunction with the increase of water temperature (Cooper et al., 1983). Accordingly, Hg transformation and cycling may be impacted by these environmental changes. Seasonal variabilities of Hg concentrations and speciation should give a better understanding of Hg cycling in this part of the system.

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Table 1. Hg concentrations (ng Hg/L) in surface water along the Carson River/Lahanton Reservoir system

Hg_T: Dissolved and particulate total mercury; Hg_{TD}: Total dissolved mercury; MeHg_T: Dissolved and particulate methylmercury; MeHg_D: Dissolved methylmercury; Hg_R: Total acid-labile Hg; Hg⁰: Elemental Hg. Values represent an average of 3 samples except when the standard deviation is not given (1 sample). Flow regimes: 546 cfs on 5-16/17-94 and 63 cfs on 6-16/17-94.

*nd; not determined; BDL: Below the detection limit.

| Locations (and Km downstream) | Collection date | Hg _T | Hg _{TD} | MeHg _T | MeHg _D | Hg _R | Hg ⁰ |
|-------------------------------------|--------------------|------------------|------------------|-------------------|-------------------|-----------------|-----------------|
| # 1 (00) | 5-16-94 | 12.88 ± 2.36 | 7.05 ± 0.43 | 0.305 | 0.161 | *nd | nd |
| | 6-16-94 | 4.28 ± 0.44 | 2.46 ± 0.51 | 0.512 ± 0.054 | 0.380 ± 0.001 | 0.46 | *BDL |
| # 2 (11) | 5-16-94 | 28.15 ± 6.30 | 3.91 ± 0.02 | nd | nd | nd | nd |
| | 6-16-94 | 7.87 ± 1.74 | 2.97 ± 1.22 | 0.505 ± 0.067 | 0.399 ± 0.029 | 0.39 | BDL |
| # 3 (28) | 5-16-94 | 133.91 ± 31.70 | 4.79 ± 0.16 | 0.542 | 0.458 | 0.10 | nd |
| | 6-16-94 | 46.95 ± 20.81 | 7.98 ± 1.59 | 1.607 ± 0.087 | 1.016 ± 0.052 | 1.13 | 0.15 |
| # 4 (44) | 5-16-94 | 722.22 ± 131.59 | nd | nd | nd | 1.67 | nd |
| | 6-16-94 | 163.41 ± 39.27 | 10.16 ± 0.36 | 2.001 ± 0.074 | 1.330 ± 0.340 | 1.95 | 0.13 |
| # 5 (69) | 5-16-94 | 2107.00 ± 59.71 | 46.09 ± 11.35 | 7.204 | 1.202 | 3.78 | nd |
| | 6-16-94 | 645.64 ± 64.24 | 9.43 ± 0.28 | 1.786 ± 0.031 | 0.844 ± 0.181 | 2.72 | 0.91 |
| # 6 (87) | 5-16-94 | 1471.00 ± 37.72 | 13.23 ± 3.63 | 4.334 | 1.597 | 2.34 | nd |
| | 6-16-94 | 824.18 ± 91.81 | 10.19 ± 1.45 | 1.813 ± 0.023 | 0.787 ± 0.029 | 3.63 | 0.17 |
| # 7 (95) | 5-16-94 | 1251.00 ± 174.81 | 56.46 ± 15.04 | nd | nd | 3.19 | nd |
| | 6-16-94 | 1582.98 ± 93.96 | 25.28 | 3.725 ± 1.090 | 0.129 ± 0.010 | 12.88 | 0.83 |
| # 8 (111) | 5-16-94 | 57.06 ± 0.77 | 3.30 ± 0.09 | 0.544 | 0.149 | 0.78 | nd |
| | 6-16-94 | 158.35 ± 15.14 | 3.98 ± 0.35 | 0.400 ± 0.033 | 0.135 ± 0.028 | 1.66 | 1.23 |

Table 2. Physiochemical parameters measured concomitantly with Hg determinations.

*BDL: below the detection limit (< 0.01 mg/L); D.O. dissolved oxygen; DOC: dissolved organic carbon; TSS: total suspended solids.

| Locations | Sampling date | Temperature (°C) | pH | H ₂ S mg/L | SO ₄ ²⁻ mg/L | Cl ⁻ mg/L | *D.O. mg/L | *DOC mg/L | *TSS mg/L |
|-----------|------------------|---------------------|------|--------------------------|---------------------------------------|-------------------------|---------------|--------------|--------------|
| # 1 | 5-16-94 | 11.7 | 7.34 | *BDL | 12.7 | 3.9 | 9.32 | 5.9 | 50 |
| | 6-16-94 | 16.0 | 8.26 | BDL | 37.0 | 8.4 | 7.78 | 6.4 | 11 |
| # 2 | 5-16-94 | 12.0 | 7.39 | BDL | 13.5 | 3.8 | 10.53 | 5.7 | 30 |
| | 6-16-94 | 19.0 | 8.33 | BDL | 35.7 | 8.3 | 8.20 | 6.2 | 4 |
| # 3 | 5-16-94 | 12.5 | 7.32 | BDL | 14.2 | 3.7 | 11.40 | 4.9 | 41 |
| | 6-16-94 | 15.0 | 8.08 | BDL | 43.3 | 9.0 | 9.03 | 5.6 | 4 |
| # 4 | 5-16-94 | 12.3 | 7.45 | BDL | 17.8 | 4.2 | 10.75 | 5.5 | 66 |
| | 6-16-94 | 18.0 | 7.96 | BDL | 89.9 | 11.5 | 9.70 | 3.5 | 3 |
| # 5 | 5-16-94 | 13.0 | 7.47 | BDL | 20.2 | 4.7 | 11.22 | 4.8 | 73 |
| | 6-16-94 | 20.0 | 8.13 | BDL | 89.8 | 12.3 | 9.50 | 2.9 | 14 |
| # 6 | 5-16-94 | 14.0 | 7.50 | BDL | 24.2 | 5.0 | 9.61 | 5.1 | 42 |
| | 6-16-94 | 21.0 | 8.57 | BDL | 83.2 | 11.8 | 10.90 | 3.0 | 17 |
| # 7 | 5-16-94 | 15.0 | 7.44 | BDL | 64.7 | 12.7 | 8.21 | 4.8 | 55 |
| | 6-16-94 | 20.0 | 8.22 | BDL | 52.6 | 11.4 | 8.50 | 4.0 | 67 |
| # 8 | 5-16-94 | 14.5 | 7.40 | BDL | 36.4 | 16.3 | 10.71 | 3.8 | 41 |
| | 6-16-94 | 21.0 | 8.64 | BDL | 43.4 | 14.5 | 9.10 | 3.6 | 51 |

Table 3. Comparison of Hg concentrations and speciation (ng Hg/L) in the Carson River/Lahonton Reservoir system with other values reported in recent literature. (1): present work; (2): taken from Gill and Bruland, 1990; (3): taken from Bloom and Effler, 1990. The abbreviations of different mercury species are spelled out in table 1. FCH = Fort Churchill.

| Aquatic systems | Hg _T | Hg _{TD} | MeHg _T | MeHg _D | Hg _R | Hg ⁰ | Pollution source |
|--|-----------------|------------------|-------------------|-------------------|-----------------|-----------------|--------------------|
| Carson River, NV ⁽¹⁾ | | | | | | | |
| Above mine tailings | 4-28 | 2 - 7 | 0.3 - 0.5 | 0.10 - 0.40 | 0.4 | - | Mining activity |
| Below mine (FCH) | 645-2107 | 9 - 46 | 1.8 - 7.2 | 0.80 - 1.20 | 2.72-3.8 | 0.91 | |
| Lahontan Reservoir | 57-1582 | 3 - 56 | 0.4 - 0.5 | 0.13 - 0.15 | 0.8-12.8 | 1.23 | |
| Davis Creek, CA ⁽²⁾ | | | | | | | |
| Above mine | 2.8 | | | | -- | -- | Mining activity |
| Below mine | 14.8 - 34 | 12.03 | -- | 2.80 | -- | -- | |
| Reservoir | 5.6 | 3.21 | -- | 2.40 | -- | -- | |
| Clear Lake, CA ⁽²⁾ | | | | | | | |
| Oaks Arm | 12 - 104.3 | 1.1 | -- | 0.28- 91.27 | 13.03 | -- | Mining activity |
| Rodman Slough | 3.61-18.05 | 1.5 | -- | 0.74- 14.21 | 3.81 | -- | |
| Lake San Antonio, CA ⁽²⁾ | | | | | | | |
| | 0.56 - 1.8 | 0.5-1.3 | -- | 0.1 - 0.5 | 0.4 - 0.8 | -- | None |
| Pyramid Lake, NV ⁽²⁾ | | | | | | | |
| | 1.94 | 0.90 | -- | BDL | 0.96 | -- | None |
| Onandaga Lake, NY ⁽³⁾ | | | | | | | |
| Range including values from surface and bottom water | 7.13/ 25.68 | 2.01/ 11.97 | 0.36/ 6.68 | 0.21/ 5.74 | 0.34/ 9.72 | 0.005/ 0.237 | Chlor-alkali plant |
| Clay Lake, ON ⁽³⁾ | | | | | | | |
| | 5 - 80 | -- | 1.8 - 2.8 | -- | -- | -- | Chlor-alkali plant |

Table 4. Pearson correlation coefficients (r) between physiochemical variables and Hg concentrations determined during the two surveys. Because of its dissimilarity, values from the Lahontan dam were excluded.

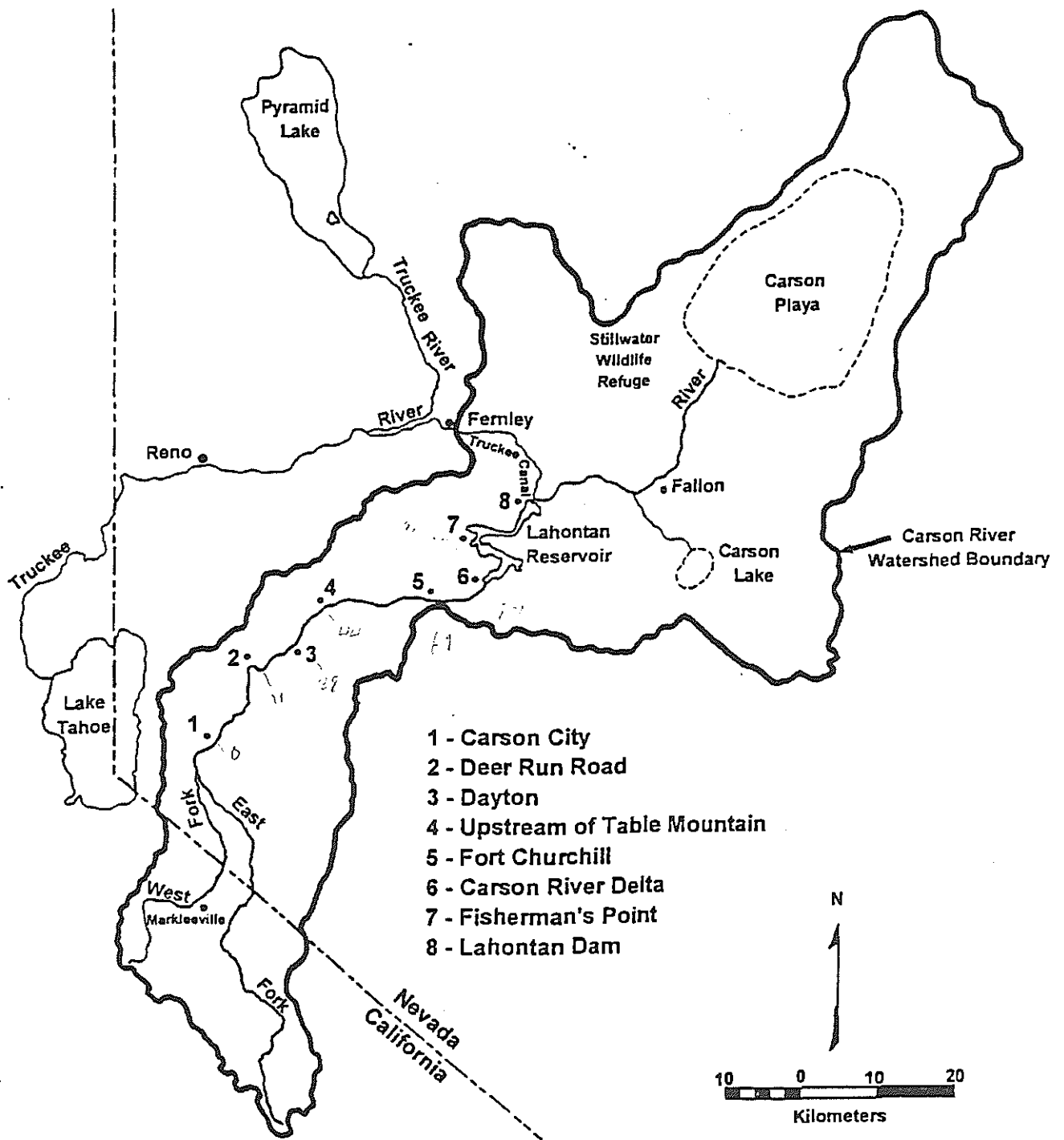
| Parameters | Hg species and concentrations | | | | |
|--------------------------|-------------------------------|------------------|-------------------|-------------------|-----------------|
| | Hg _T | Hg _{TD} | MeHg _T | MeHg _D | Hg _R |
| Distance downstream (Km) | 0.81** | 0.51 | 0.67* | 0.41 | 0.71* |
| Dissolved Oxygen | 0.34 | 0.39 | 0.36 | 0.36 | -0.09 |
| Temperature | 0.05 | -0.14 | -0.09 | -0.10 | 0.48 |
| TSS | 0.73* | 0.72* | 0.64* | -0.13 | 0.51 |
| DOC | -0.35 | -0.20 | -0.25 | -0.29 | -0.44 |
| Sulfate | -0.05 | -0.15 | -0.09 | 0.21 | 0.25 |
| Chloride | -0.05 | -0.16 | -0.12 | 0.01 | 0.42 |
| pH | -0.15 | -0.27 | -0.09 | -0.10 | 0.27 |

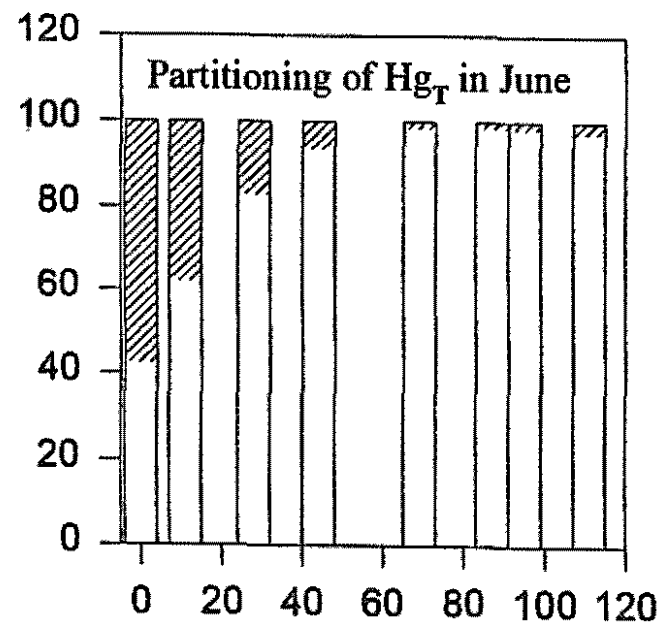
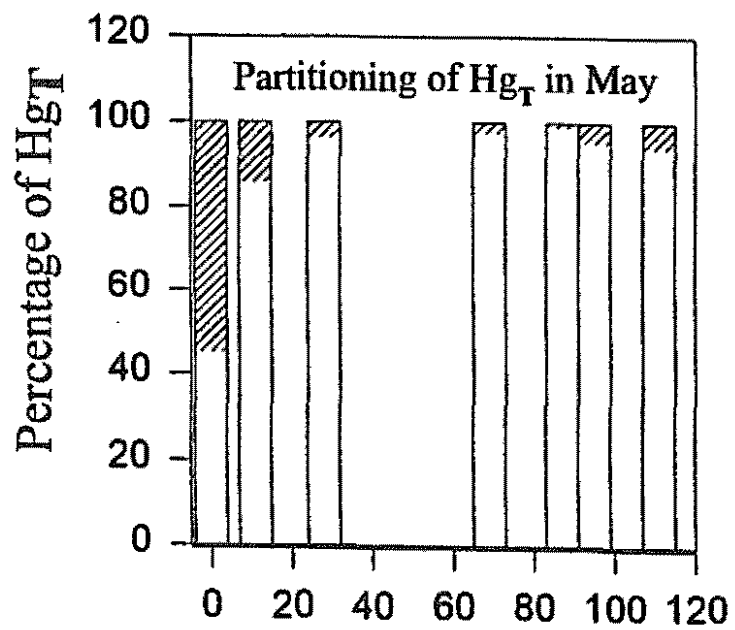
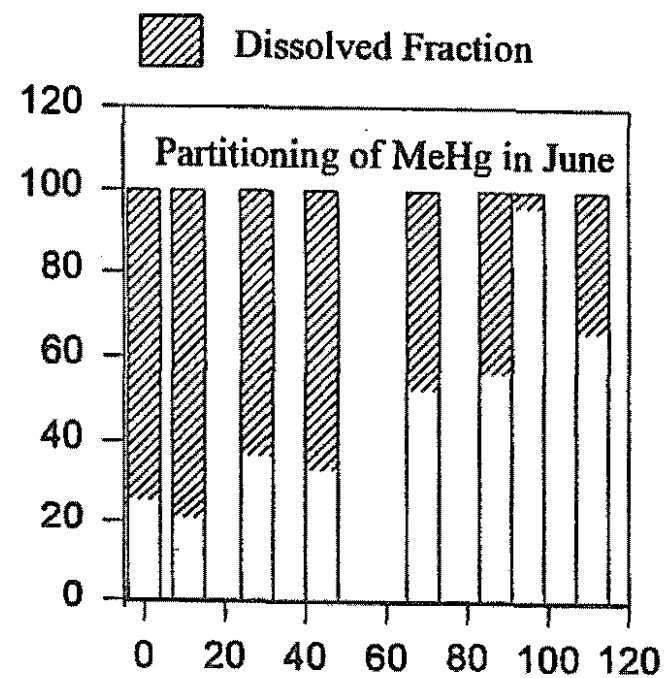
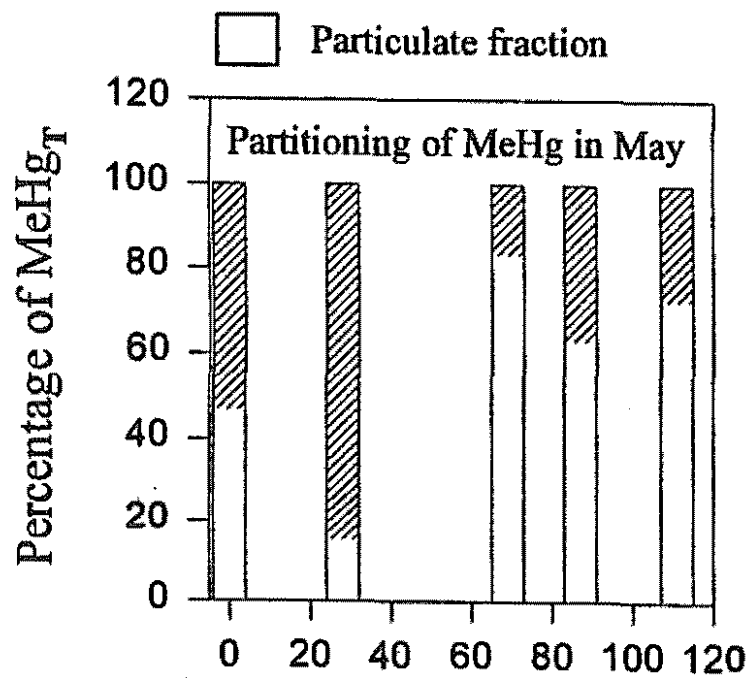
Statistical significance of correlation coefficients indicated at the 0.05(*) and 0.01(**) levels.

FIGURE CAPTIONS

Fig. 1. Location of sampling sites along the Carson River.

Fig. 2. Partitioning of total mercury (Hg_T) and methylmercury (MeHg_T) during the two sampling periods.





Distance downstream from Carson City Gage (Km)